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ARRANGEMENT AND UP-TO-DATE RESULTS OF THE SAMPLING MEASURING STATION FRANKFURT/MAIN

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Arrangement and Up-to-date Results of the Sampling Measuring Station Frankfurt/Main [Einrichtung und bisherige Ergebnisse der Probemessstation Frankfurt/Main]

by

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Introduction

Continuous recording stations for monitoring the degree of outside air pollution to date have only been built in the United States. There, by order of the Department of Health, Education and Welfare, the Center for Air Pollution Control operates such a station in six large cities. Continuous recording measuring devices are used for the detection of the outside air concentration of hydrocarbons SO_2, NO_2, NO, CO and oxydants. The measurements began in 1962; the first results could be published in

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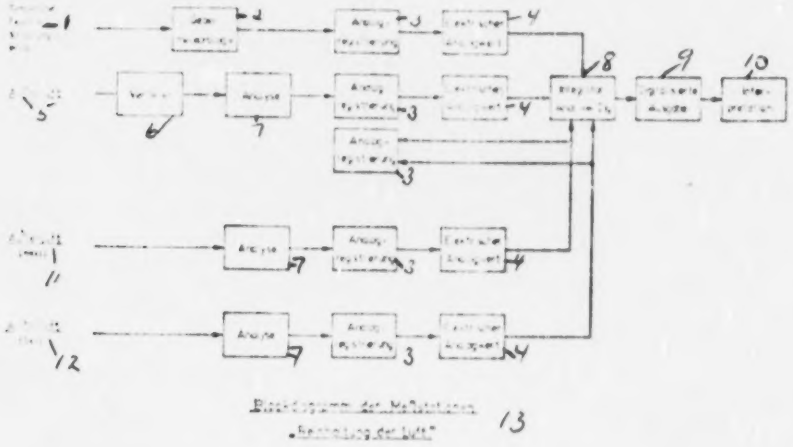
1965 [1]. The program was initiated through a federal law [2]; Congress directed the U. S. Public Health Service to investigate the effects of motor vehicle exhaust gas on human health. The Continuous Air Monitoring Program (CAMP) was supposed to monitor the concentration of different gaseous air contaminants of the atmosphere and to examine harmful effects on man, animal, plants and inanimate objects.

As an extension of this intent and under consideration of the knowledge gained there, the Bundesregierung (Federal Government of West Germany) has issued a law regarding preventive measures for maintaining clean air [3]. The density of the concentration of different toxic air-polluting substances is supposed to be continuously recorded. Simultaneously, the meteorologically limiting quantities are supposed to be available, which are necessary for the description of the quality air situation. Like the new United States regulations, the goal is to obtain information about necessary remedial and preventive measures besides determining the degree of pollution. The U. S. Public Health Service [4], under consideration of the actual status of the air pollution and an estimation of the changes in the next ten years, already gave particulars out of the measurements of the CAMP about the necessary percental reduction of the emissions.

Only through the simultaneous recording of the meteorologically limiting quantities with the aid of many years of a series of recordings of the German Weather Bureau, it was possible to make a statistically guaranteed forecast about the development and degree of air pollution. In the Ruhr region research has shown during the last years that despite the constant emissions, displacements in the level of the immissions could

occur. Through the simultaneous detection of meteorological factors, for example, through the calculation of diffusion classes [5], we can assign the immission concentrations to certain propagation conditions. Thus the decision can be made of whether a change of the emissions or a changed weather condition is responsible for this. An emission cadastre, as already exists in a few Bundeslaender (counties), is, however, an absolute prerequisite for such considerations.



Arrangement of the Sampling Measuring Station Frankfurt/Main (Ill. 1)

Key:
1. Temperature, moisture, radiation, wind
2. Transmitter, meteorology
3. Analogous recording
4. Electric analog values
5. Outside air
6. Distributor
7. Analysis
8. Integrator, analog.--digital
9. Digital output
10. Interpretation
11. Outside air (West)
12. Outside air (East)

13. Block diagram of the measuring stations "Clean Air Maintenance"

The technical design was executed similar to that of the CAMP-stations of the U. S. Health Service [6]. Through a ring conduit to which the analysis devices with their intake pipes are connected, the air is sucked through with a velocity of flow of approximately 1 m/sec (2.5 m^3/h).

For the comparability of the concentration changes of different components, it is necessary that all devices indicate approximately the same transient response. Therefore, intake pipes and pumps of the individual analysis devices are laid out in such a manner that a concentration change appearing on one of the joint suction places can be recorded by all devices after 20 - 25 sec. through the beginning increase of the analogous recording. With uniform temporary duration of the concentration increase, according to our experiments all devices start also approximately simultaneously after 35 - 40 sec. with the indication of the concentration decrease. Therefore, the basis of this is a uniform synchronism of the analysis devices.

LARSEN, BENSON and JUTZE [7] examined the intake function of different analysis equipment. If C_t represents the true outside air concentration, C_i the concentration recorded by the analysis device, C_1 the true outside air concentration before the start of the concentration change, t the time after the appearance of the change and p an equipment constant, then

$$(C_t - C_i) = (C_t - C_1)\exp(-t/p) \qquad (1)$$

and for t = p

$$C_t = C_i + p\frac{dC_i}{dt} \qquad (2)$$

After the time interval p, the difference ($C_t - C_i$) between the true and the recorded concentration has dropped to 36.8 % of the original difference. According to equation 2, the true concentration can be computed when the recorded concentration and the transition function $p = \frac{dC_i}{dt}$ are known.

The time constant of our analysis devices is approximately 30 sec. With certain experiments, concentration differences must still be readable with time steps of five minutes; at the same time, the differences $C_i - C_1$, for example, with SO_2, amount to a maximum of approximately 0.05 ppm. If C_t is the true concentration between the time point t_1 (with concentration C_2 = 0.07 ppm), then

$$C_t = \frac{C_1 + C_2}{2} + \frac{p\,(C_2 - C_1)}{t_2 - t_1} = \frac{0.09}{2} + \frac{0.5 \times 0.05}{5}$$

$$C_t = 0.050 \text{ ppm } SO_2$$

compared to an instrument reading of 0.045 ppm SO_2. It could, however, be shown that the true concentration at individual time points deviates considerably from the one recorded, evoked, as a rule, through a time lag behind the true course of the recorded concentration so that, consequently, the course of both concentrations is only time displaced, otherwise, however, parallel and with almost equal amplitude. According to these calculations for air quality experiments, the time constants of the devices should be approximately half as long as the shortest time span that is intended for averaging. With a time constant of only 30 sec., this demand is very well met with the equipment of the sampling measuring station.

The construction of the sampling measuring station was described by

SCHUETZ [8]. For monitoring the SO_2 concentration, SO_2 analyzers were placed into the three stations. In addition to this, a CO_2 recorder had to be installed for emitter analysis, that is, analysis of room heaters, motor vehicles, chemistry. Within a reasonable space of time, the continuous detection of the NO_2 concentration will be added at each of the three stations in order to gain a more exact coordination of the emissions from exhaust gases and, furthermore, to be able to estimate the effect of motor vehicle traffic at outlying stations.

Besides the recording of CO_2 and SO_2, the recording of the carbon monoxide concentration and the total hydrocarbon content (HC) of the outside air is available in the central measuring station. Another NO_2 analyzer can be put into service here later. Together with measurements of wind velocity and wind direction, temperature, moisture and radiation balance, quite extensive measurements of the parameter of quality air research are possible. It will be especially evident whether we can dispense with the one or the other recording in the future without decreasing the prediction power.

Compared to CAMP, the form of the conversion from an analog recording in digital values had to be improved. The first CAMP station was planned at the end of the 1950s; tape reader punching was used, which, at intervals of five minutes, punched each of the instantaneous values off the analyzing devices. The formation of half-hour averages from these six instantaneous values certainly is quite dependable for statistical analysis; deviations of individual cases are cancelled in the devices. However, we have the intention to examine the chronological sequence of the concentration pattern and perhaps to consider several air pollution

materials side by side. The questioning and the recording of the five-minute instantaneous values are no longer adequate for this, therefore, a balance computer for the digitizing has been installed, which is in the position to compute three-, six- and thirty-minute averages. Thus it is also possible, for example, to track in a chronological course the three-minute averages of two parallel recordings of different substances and to relate them to each other. In addition, the time constants of our equipment amount only to approximately 30 sec. In the CAMP stations, equipment was used in part whose time constants amounted up to ten minutes [7]. Through the short time constants of our analyzers, through the development of real averages from the analog recordings and through the almost uniform excess function of all analyzers during concentration changes, the prerequisites for a comparable analysis have been produced by us for the sampling measurement station.

Site of the Measuring Station

The most important task of the immissions measuring is the production of data for improvement measures. Through the continuous recording of different components of air pollution and the meteorological factors in the sampling measuring station Frankfurt/Main, we have taken the first steps towards an emitter analysis. It will be possible within established limits to determine the percentage of immissions from heaters, power plants, chemical factories and motor vehicles. Through the installation of several such recording stations at selected sites within an agglomeration area, this prediction can still be improved. Such programs were not carried out in the past. We are in the position to recognize this possibility from

the research in Nashville [9], New York [10] and Readings [11]. In Nashville and New York, a predominant influence of the high emission rates of the city centers affected the level of the SO_2 concentration. With east winds in Readings under the influence of the descending air current plume from Greater London (at a distance of approximately 50 km), maximum SO_2 concentrations were found. An examination of the horizontal distribution showed that basically with all weather conditions, the highest values occur in the center. In New York, besides the SO_2 concentration, the CO concentration was also examined at several measuring sites. The result was an interrelation, discovered also by other authors [12], [13], between the CO concentration, the distance of the measuring place from the street and the traffic frequency. The strain on the

Radius (m)	r	Radius (m)	r
250	0,72	1750	0,86
500	0,76	2000	0,86
750	0,77	2250	0,85
1000	0,84	2500	0,83
1250	0,84	2750	0,82
1500	0,85	3000	0,80

Table 1

metropolitan city air through auto exhaust gases could be represented quite well through the combined recording of SO_2 and CO with the aid of the meteorological test values.

Research in Readings [11] proved furthermore that a correlation exists between the degree of the SO_2 emission in the vicinity of a measuring

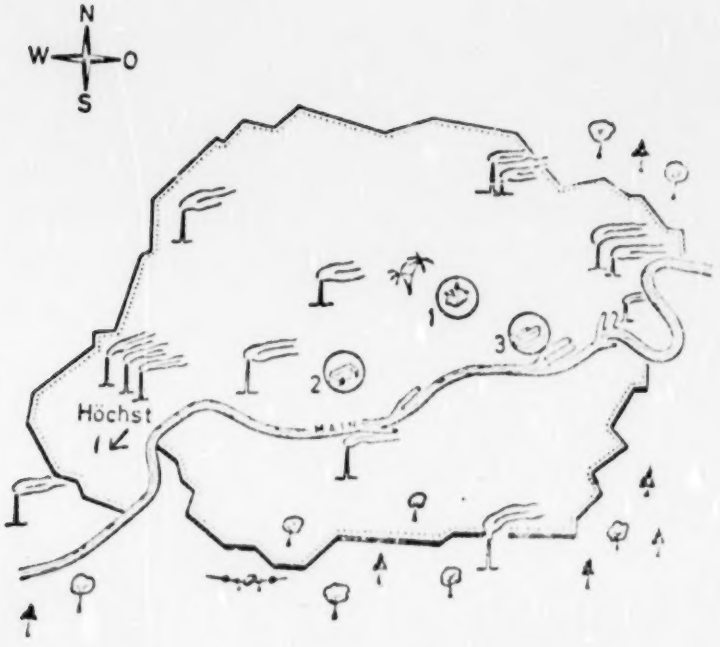


Illustration 2

Key: 1. Highest

station and the degree of the SO_2 immission. The correlation (r) between the SO_2 concentration and the SO_2 emission was calculated for ever-increasing radii around the station. Thus the following values resulted (see Table 1):

At a distance greater than 3000 m, however, the correlation coefficient, of course, drops fast.

Research in Nashville [14] resulted in an excellent consistency of the frequency distributions, which were obtained from different dense measuring grids. In the winter the variations of the two frequency distributions from one measuring grid with 32 stations (in an area of approximately

180 km^2), with seven stations and with only two stations (one station in the center and another station at the edge of the city, distance 5.5 km) amounted to:

It is clearly evident that in the winter with larger measuring grids, approximately 30 % higher SO_2 concentrations have been ascertained.

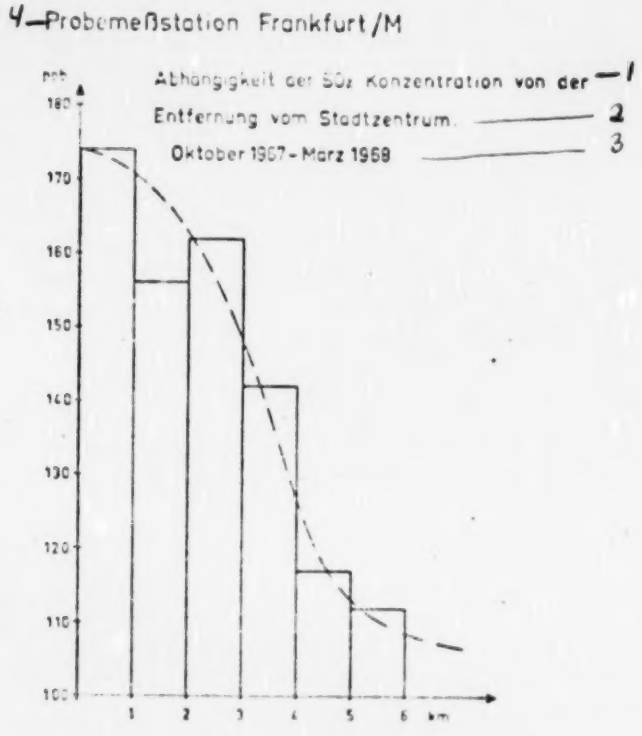


Illustration 3

Key: 1. SO_2 concentration subject to
2. distance from the city center
3. October 1967 - March 1968
4. Sampling measuring station Frankfurt/Main

With the experiments with one measuring grid consisting of two stations, the station at the edge of the town was installed too far outside.

The concentration at this site was only immaterially higher than the background concentration outside of the city [15]. On the basis of the horizontal distribution of the SO_2 concentration, an average SO_2 concentration was supposed to be present, which, however, lies in the center between the maximum concentration in the city center and that outside of the city. The distance between the two selected stations then should have only amounted to approximately 4 km, if one station is located in the center on the site of the highest concentration.

Basically, experiments of the horizontal distribution show, however, that generally in the three cities mentioned, the maximum concentrations can be found in the center. This evidence is always valid for the heating period, which in our latitudes is accompanied by physiologically dangerous concentrations. In the other annual latitudes, this evidence about the horizontal distribution can naturally become obscured through industrial influences.

The sampling measuring station Frankfurt/Main (Illustration 2) was built on the basis of these results. For technical reasons, of course, the central measuring station could not be installed directly in the center of Frankfurt; it lies approximately 1200 m from the actual center. If we assume approximately similar conditions in Nashville [14], then the concentration at this site should lie approximately 10 % lower than at the maximum point. In the west a measuring site is located at a distance of 4 km, in the east at 2 km from the center. The distance from the central station to the west station and to the east station is 3.5 km.

The Measuring and Inspection Office of the Industrial Control Administration in Hessen in a separate test determined the horizontal

distribution of the SO_2 concentration in a circular area with a radius of 3 km around the respective station. Available here are only the measurements from October 1967 to March 1968. Illustration 3 thows the SO_2 concentration subject to the distance from the city center. Similar to the Nashville program [15], a reduction function occurs, which has the form of a normal distribution. The normal distribution can be represented in the form

$$y = \frac{\exp[-1/2\,(x\,s)^2]}{2\pi} \qquad (3)$$

Here $x = x_1 - x$, that is, the value of the variables, is relative to the distance from the average and s the standard deviation.

If S is designated as the SO_2 concentration, S_b the SO_2 concentration of the so-called "background" and S_c the concentration in the city center and r the distance of the measuring site from the city center, the equation for the SO_2 ground concentration subject to the distance from the city center is obtained in the form of

$$S = S_b + S_c \exp[-1/2\,(r/s)^2] \,. \qquad (4)$$

The location for our stations for detecting the SO_2 horizontal distribution is therefore more favorable as were the two chosen stations in Nashville [14]. The central station is located at a site where the SO_2 content amounts to approximately 94 %, the east station at a site where the concentration amounts to approximately 87 % and the west station at a site where the SO_2 concentration amounts to approximately 40 % of the city center. The background concentration was assumed to be 100 ppb SO_2, which is admissable for the winter months and the densely populated Rhein-Main area.

A comparison of the frequency distribution of the SO_2 concentrations, measured once with the three continuously recording sampling measuring stations and measured otherwise at the 66 measuring points of the grid according to TAL, results in the following values:

Wert der Summenhäufigkeitsverteilung % (1)	Alle Stationen (2)		Weststation (3)		Zentralstation (4)		Oststation (5)	
	a	b	a	b	a	b	a	b
50	103	130	102	108	113	138	85	138
80	182	215	222	183	182	240	152	225
90	242	270	308	260	235	290	182	275
95	304	320	375	278	297	425	245	425
97,5	355	365	415	290	310	470	340	450
99	425	430	470	296	395	500	335	500

a = Werte der Probemeßstation, b = Werte nach der TAL (6)

Table 3

Key: 1. Value of accumulative frequency distribution %
2. All stations
3. West station
4. Central station
5. East station
6. a = values of the sampling measuring station, b = values according to TAL

The consistency of the measuring values of the grid from the three continuously recording stations with measurements at the 66 measuring points according to TAL is practically complete. The individual stations and the values of the respective measuring area naturally still show deviations because of the small number of the measuring value.

But the consistency is unmistakenly better than in Nashville (Table 2). As mentioned above, this probably is attributed to the more favorable

selection of the measuring locations.

Wert der Summenhäufigkeitsverteilung 1	32 Stationen ppb SO_2 2	7 Stationen ppb SO_2 3	2 Stationen ppb SO_2 4
10 %	90	80	65
50 %	300	250	220
80 %	600	550	500
90 %	1000	800	750
Abstand zwischen den Stationen etwa 5	3,5 km	5,5 km	5,5 km

Table 2

Key: 1. Value of the cumulative frequency distribution
2. 32 stations ppb SO_2
3. 7 stations ppb SO_2
4. 2 stations ppb SO_2
5. Approximate distance between the stations

If we incorporate the results of Readings [11] and assume that within a radius of 3 km around the respective station a correlation coefficient still exists of 0.8 between the degree of emission and the immission measured at the station, then the measuring results of these three stations would still be valid for almost 80 % of the built-up area of the city of Frankfurt. This means that we are in the position to:

1. ascertain with the respective station the temporary changes for the circle with a radius of 3 km and

2. maintain the absolute concentrations with the aid of a measurement of the horizontal distribution taken once and with the aid of a test of the concentrations subject to wind vectors or diffusion classes.

Combination Analysis

The sampling measuring station was the result of the recognition that in order to control the air pollution and to estimate the noxious secondary effect, characteristic substances must be continuously tested and, if possible, without interruption, for several years. An estimation about the level of emission in the area of Nashville has been taken from the research of SCHUENEMAN, WILLIAMS and EDMISTON [16]:

(Angaben in tons per year) 1

	Komponente 2	Gasfeuerung 3	Kohlefeuerung 4	Heizölfeuerung 5	Kfz-Benzin 6	Kfz-Diesel 7	Müllverbrennung 8	Chemische Industrie 9
10	Aldehyde	283	—	23	311	9	169	—
11	$NO + NO_2$	2542	1387	710	6225	328	25	21
12	Organische Gase	51	1787	23	18800	328	11914	1755
13	Schwefeloxyde	12	28950	882	459	82	50	?
14	Feste Stoffe	—	6789	134	685	229	1982	1384

Table 4

Key:
1. Data in tons per year
2. Components
3. Gas heating
4. Coal heating
5. Heating oil burning
6. Motor vehicle gasoline
7. Motor vehicle Diesel
8. Garbage burning
9. Chemical industry
10. Aldehyde
11. $NO + NO_2$
12. Organic gases
13. Sulfur oxide
14. Solid materials

Naturally, this table is only valid for the emitter composition as it exists in Nashville. It is furthermore incomplete since data about the CO emissions (which mainly must be attributed to exhaust gases of motor vehicles with Otto carburetor engines [17]) and the CO_2 emissions (whose

main contributions are furnace installations) are missing. The small emission of organic gases during the burning of heating oil is only then valid when the installations function in an optimum situation. This is rarely the case with older installations or those that have been changed from coal to oil.

For research in regard to an emitter analysis through a combined analysis of immission measurements, the values represented in Table 4 can be summerized somewhat different.

	Hauptemissionen 1	weniger wichtige Emissionen 2
1. Feuerungsanlagen: 3		4
a) Hausbrand und Zentralheizungen	SO_2, CO_2, NO_2	organische Gase, Aldehyde, CO
b) Kraftwerke	SO_2, CO_2, NO_2 (Kohle: organische Gase) 6	Aldehyde, CO 7 (Heizöl: organische Gase) 8
c) Gasfeuerung	Aldehyde, CO_2, NO_2	organische Gase, SO_2, CO
2. Kraftfahrzeuge 5	CO, NO, organische Gase 9	SO_2, CO_2, Aldehyde 7
3. Müllverbrennung 10	organische Gase 9 (NO_2), CO_2	7 Aldehyde, (NO_2), SO_2, CO
4. Chemie 11	organische Gase 9 (eventuell SO_2), 12 (eventuell NO_2) 13	7 Aldehyde, CO_2, CO

Table 5

Key:
1. Main emissions
2. Less important emissions
3. Furnace installations
   a) domestic fuel and central heating
   b) power plants
   c) gas heating
4. Organic gases, Aldehyde, CO
5. Motor vehicles
6. (Coal: organic gases) aldehyde, CO_2, NO_2
7. Aldehyde, CO
8. (Heating oil: organic gases) organic gases, SO_2. CO
9. Organic gases

10. Garbage burning
11. Chemistry
12. (Perhaps SO_2)
13. (Perhaps NO_2)

Through steady parallel recording of the concentration of selected components under consideration of the wind direction and wind velocity, a prediction about the origin of the air pollution can be made.

A few examples are listed in the following Table 6. Parallel development of the concentration of the components represented in column 1 and missing parallelism to the gases in column 2 led to the emitter data in column 3.

	paralleler Verlauf 1	unbeeinflußt 2	Emittent 3
1.	$CO + NO_2 + HC$	SO_2, CO_2	Kfz 4
2.	$SO_2 + CO_2 + NO_2$	CO, HC	Feuerungen Öl oder Gas } 5
	$SO_2 + CO_2 + NO_2 + HC$	CO	Feuerungen Kohle } 6
3.	SO_2	alle anderen 7	Chemie 8
	NO_2	alle anderen 7	Chemie 8
	HC	alle anderen 7	Chemie 8

Table 6

Key: 1. Parallel development
2. Unaffected
3. Emitter
4. Motor vehicle
5. Heating oil or gas
6. Heating coal
7. All others
8. Chemistry

Because of the measurement-technique reasons, the immission measurement must remain limited to a few components while many more substances can be analytically detected with emission tests. If an emitter group

can be determined from the recording of the sampling measurement station, the approximate concentration of materials in the outside air and not detected by us can be computed with the aid of emission and immission analyses. If, for example, aldehyde and nitric oxide are represented with gas heating, approximately at a ratio of 1:10, then it can be assumed that this ratio will also occur at the place of immission. If it is ascertained that the pollution stems from gas-fired installations and if we measure the NO_2 concentration of, for example, 200 ppb at the immission site, then the aldehyde content of the exhaust gas plume must amount to approximately 20 ppb. Such considerations are limited where one or the two components are subject to a change during the duration in the atmosphere. The influence of sun irradiation during the smog formation in Los Angeles leads to a change of the relationship between the nitric oxide, carbon monoxide and hydrocarbons and simultaneously to a regeneration of oxidants, which originally were not present in the emissions [18]. On the other hand, improvements of the air quality can be expected when SO_2 will become oxidized, adsorbed or absorbed [19]. Outside of the agglomeration areas, the SO_2 concentration very quickly drops to low values [20]; the average duration in the lower troposphere was determined at approximately four to six days.

Emitter Analysis

These considerations of a combination analysis have first been used on those immissions which we assumed had been caused by exhaust gases from heating installations, either room heaters or power plants. These cases can be easily recognized in the recordings: with an unchanged

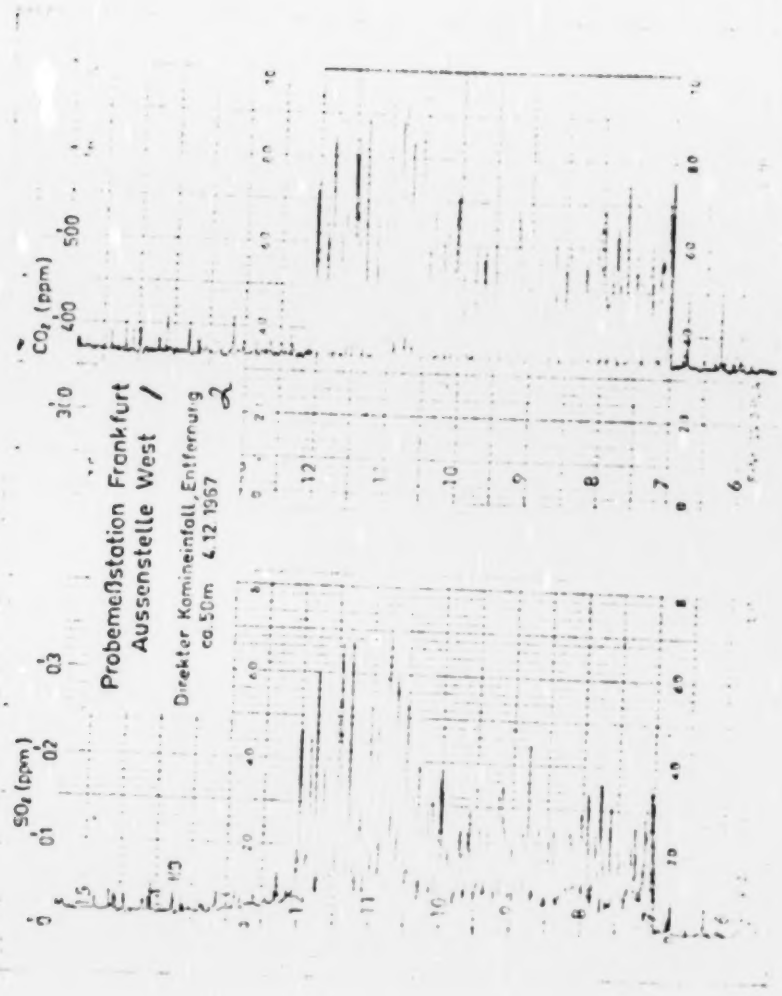


Illustration 4a

Key: 1. Sampling measuring station Frankfurt/Main, outlying station west
2. Direct fall from chimneys, distance ca. 50 m. 11-4-67

concentration of CO and HC, the recordings of SO_2 and CO_2 must indicate a parallel course. We have investigated the time span May 1967 to March 1968 as much as possible for characteristic and well-defined cases and and have determined that mainly three types of concentration changes occur:

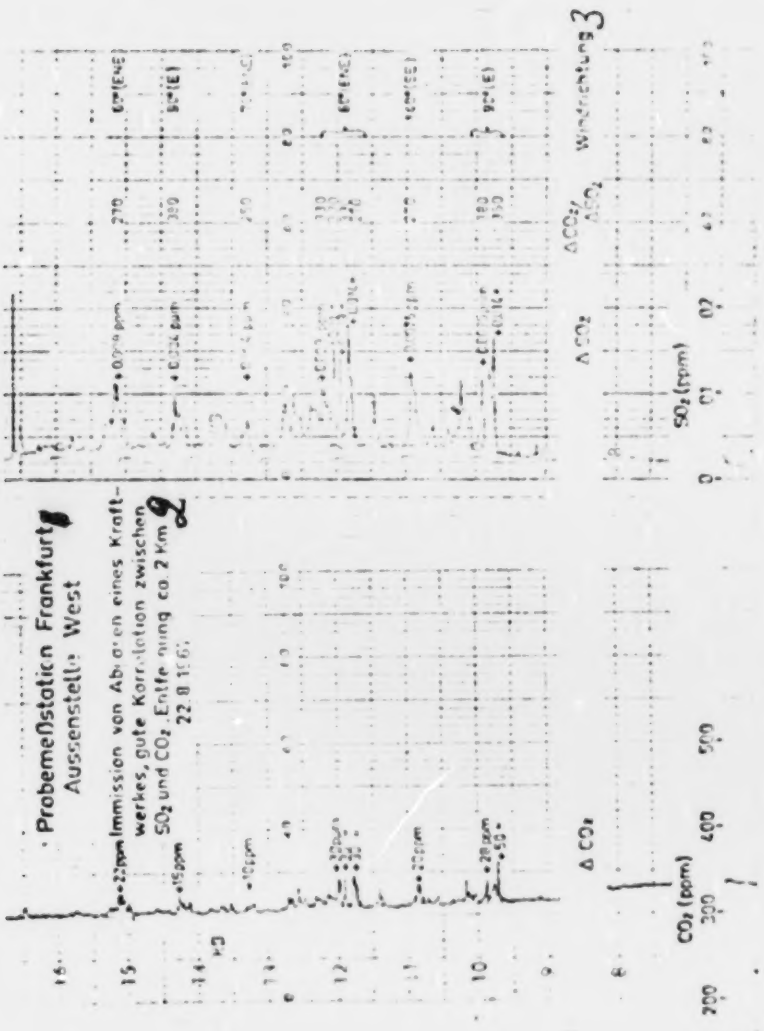


Illustration 4b

Key: 1. Sample measuring station Frankfurt/Main, outlying station west
2. Immission of exhaust gases of a power plant, good correlation between SO_2 and CO_2, distance ca. 2 km, 8-22-67

1. Short periodic changes with a period of approximately 1 - 5 minutes (Illustration 4)

2. Concentration changes with a period of approximately 10 - 30 minutes (Illustration 5)

3. Slow changes of the SO_2 and CO_2 concentrations; duration of an increase up to a few hours (Illustration 6)

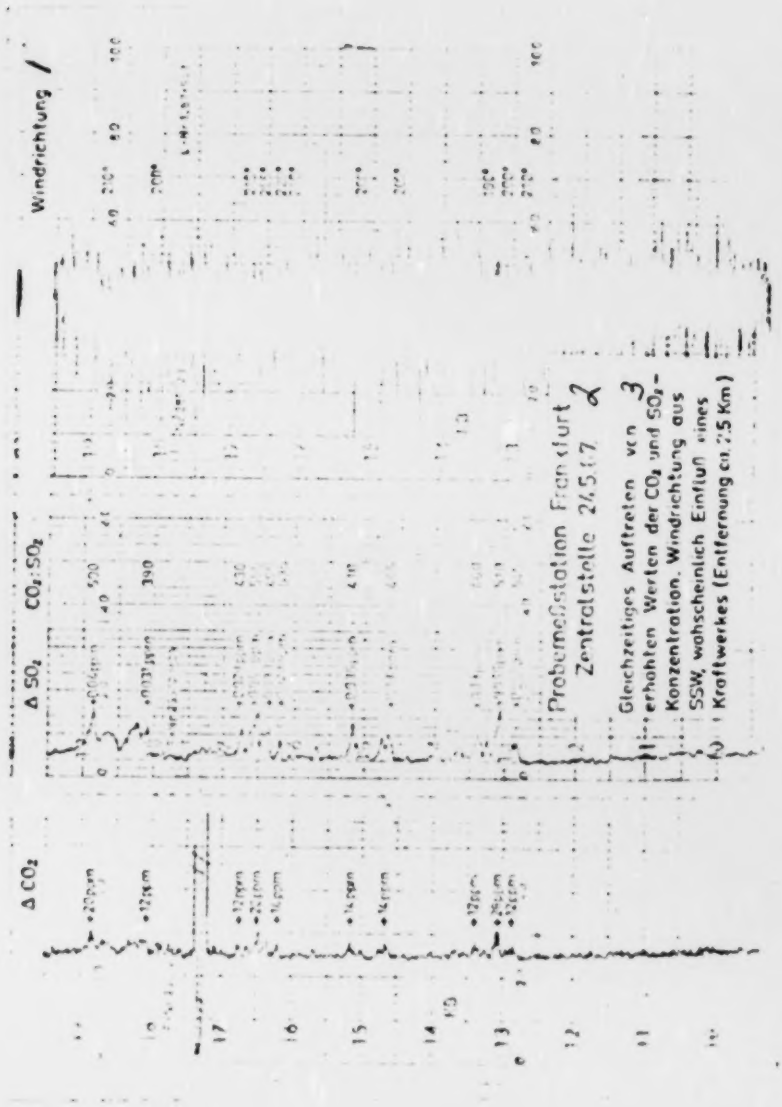


Illustration 5a

Key: 1. Wind direction

2. Sampling measuring station Frankfurt/Main, central site, 5-24-67
3. Simultaneous appearance of increased values of CO_2 and SO_2 concentration. Wind direction from SSW; probably effect of a power plant (distance ca. 2.5 km)

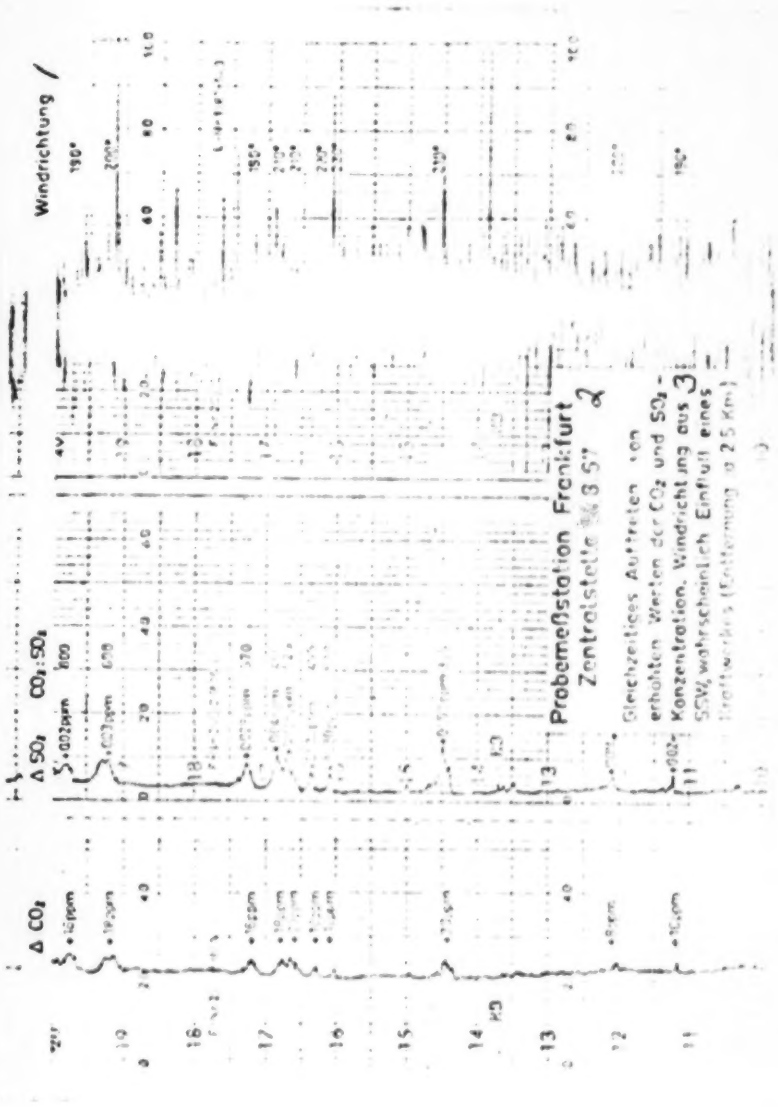


Illustration 5b

Key: 1. Wind direction
2. Sampling measuring station Frankfurt/Main, central station, 8-14-67
3. Simultaneous appearance of increased values of CO_2 and SO_2 concentration, Wind direction from SSW; probably effect of a power plant (distance ca. 2.5 km)

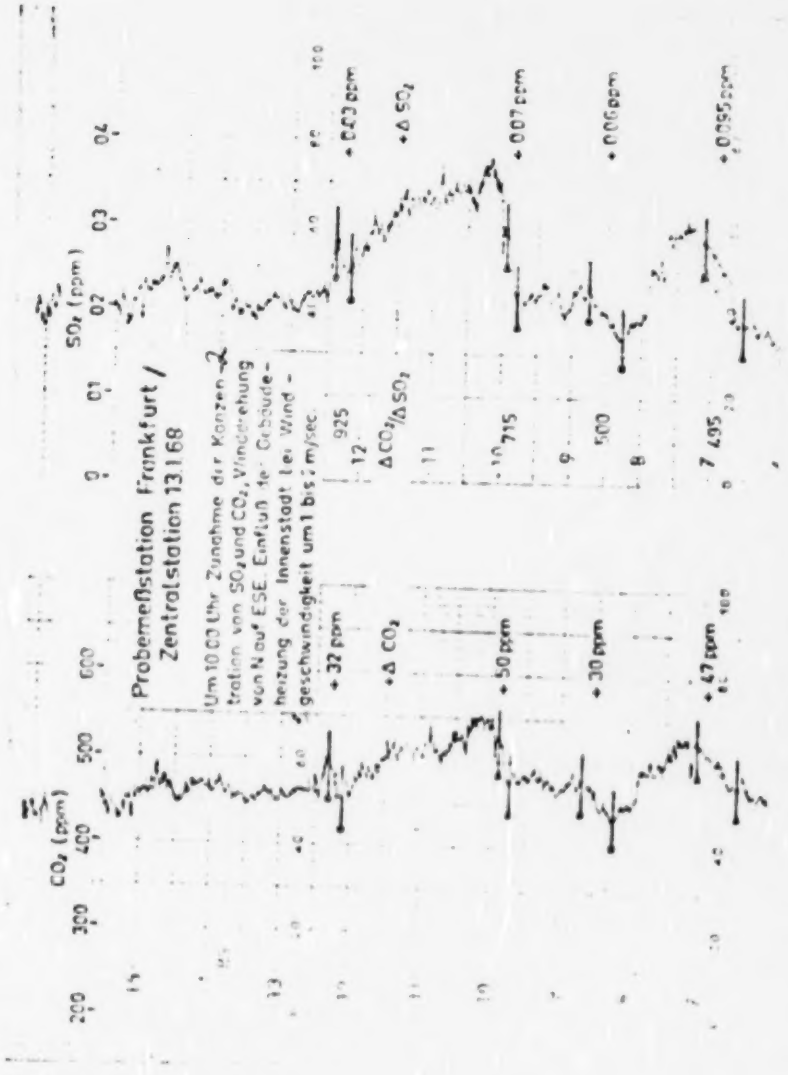


Illustration 6a

Key: 1. Sampling measuring station Frankfurt/Main, central station 1-13-68

2. At 10:00 a.m. increase of concentration SO_2 and CO_2, change in wind from N to ESE. Influence of the building heaters of the central town at wind velocity from 1 to 2 m/sec.

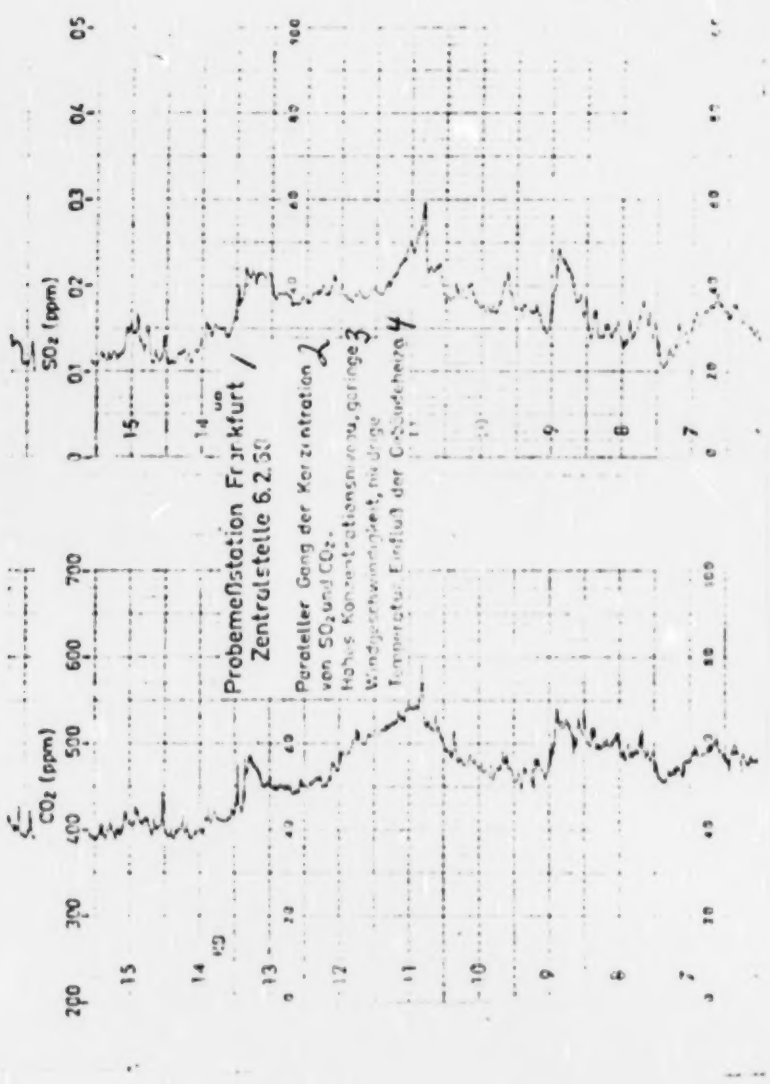


Illustration 6b

Key: 1. Sampling measuring station Frankfurt/Main, central station 2-6-68
2. Parallel course of concentration SO_2 and CO_2
3. High concentration level, low wind velocity
4. Low temperature, influence of building heaters

The physical appearance according to Illustration 4 can be dispensed with for further analysis. According to our observations, it concerns influences of very close emissions, approximately at a distance of 50 - 100 m from chimneys of neighboring buildings. Concentration changes according to Illustration 5 must still be analyzed through the observation of wind conditions. We believe that this shape of parallel SO_2 and CO_2 changes concerns the effects of plume-shaped exhaust gas concentrations of large emitters. As can be easily recognized from Illustration 7, these cases in the sampling measuring station, especially in the summer, could be primarily observed with wind directions from the sector south to west. Power plants are located in this sector in direction SWS, also in direction WSW.

During the colder time of the year, the observed case is clearly lower according to Illustration 5; the correlation of the wind direction is hardly recognizable. Through the influence of strong emissions of the heater installations, the SO_2 concentration is higher and in addition, is subject to strong variations. The distribution of the SO_2 concentrations according to earlier tests [21], for example, in the summer, amounts to only 47 $\mu g/m^3$ with an average of 55 $\mu g/m^3$, in the winter, however, to 187 $\mu g/m^3$ with an average of 334 $\mu g/m^3$. Thus exhaust gas plumes from large-scale emitters fall down on the measuring site and in many cases are concealed through other influences. Only on days with a relatively even course of the SO_2 concentration, can the superimposed concentration maxima

of the immissions by the large-scale emitters still be determined. Also, in areas where the influences of room heaters is small, an examination of the influences of the large-scale emitters can be carried out with success. From the representation of the CO_2 and SO_2 maxima (Illustration 7), it can be recognized that by far the most cases appear in the sector south to west. There two large power plants are located at distances of 2.5 km and 10 km with a maximum discharge of up to 3000 kg of SO_2 per hour each. Approximately, in the north direction lies the central garbage burning installation of the city of Frankfurt; in the sector toward NE, a central heating system (also in operation during the summer) is located at about 1 km distance (Illustration 2).

If we combine the cases from the sector south to southwest (I) (power plant with coal firing at a distance of 2.5 km) and southwest to west (II) (power plant with coal and oil burning at a distance of 10 km) and those from the remaining sectors, the following representation results:

Windrichtung 1	$CO_2 : SO_2$	Zahl der Fälle 2
I: 180°—225°	476	48
II: 225°—270°	609	25
III: 270°—360° 360°—180°	1288	26

Table 7

Key: 1. Wind direction
2. Number of cases

At the same time, we know from the analyses of the composition of the emissions that certain emitter groups have typical CO_2:SO_2 ratios. The

representation of BREUER (22) offers the following:

Brennmaterial 1	S-Gehalt 2	CO_2:CO_2 (Emission) 3	
4 Benzin	0,05 %	2500	
5 Heizöl EL	0,3 %	400—500	Heizungsanlagen 9
6 Koks, Kohle	1,0—1,2 %	200—300	Heizungsanlagen 9
7 Heizöl S	etwa 2,1 %	100	Kraftwerk 10
8 Pechkohle	4,5 %	54	Kraftwerk 10

Table 8

Key: 1. Burning material
2. S content
3. CO_2:CO_2 (emission)
4. Gasoline
5. Heating oil EL
6. Coke, coal
7. Heating oil S
8. Bituminous coal
9. Heater installations
10. Power plant

The values gained from the immission measurements of the sampling measuring station lie considerably higher than the emission ratios of Table 8. This is still more clearly expressed when we consider the quantity distribution of the cases from May 1967 to September 1967.

The testing time period lies outside the heating period. Most of the cases occur with wind directions from the sector with the two large power plants (Illustration 7). Only distinct and exactly parallel rises of SO_2 and CO_2 concentrations were evaluated (Illustration 5). We therefore have reason to assume that all the cases itemized in Table 9 were caused by

such emissions. Nevertheless, the characteristic value for the CO_2:SO_2 ratio for power plant emissions occurred only in approximately 20 % of all cases.

CO_2:SO_2	Weststation % 1	Zentralstation % 2	Oststation % 3
100—200	1,7	1,3	3,1
200—300	14,5	5,6	6,2
300—400	21,5	19,0	22,0
400—500	19,0	17,0	23,0
500—600	21,5	15,5	37,0
600—700	9,7	14,5	22,0
700—800	4,5	5,6	7,0
800—900	2,1	5,6	7,5
Zahl der Fälle 4	(271)	(360)	(154)
Mittelwert 5	500	660	900

Table 9

Key: 1. West station
2. Central station
3. East station
4. Number of cases
5. Average

Since the CO_2 content in a smoke plume with certainty experiences no change besides dilution with the atmosphere, the SO_2 portion must have changed between the chimney and the measuring place. While the average duration of sulfur dioxide in the lower troposphere is reported to be four to six days, MEETHAM [23] already inferred earlier that this value is not valid for heavily polluted areas. He calculated an average duration of only a few hours for the agglomeration area in central England. According to the results in Table 9, this time span could even be shorter, which we, however, still have to examine more thoroughly.

As could be expected, the result is that the relative SO_2 loss in an

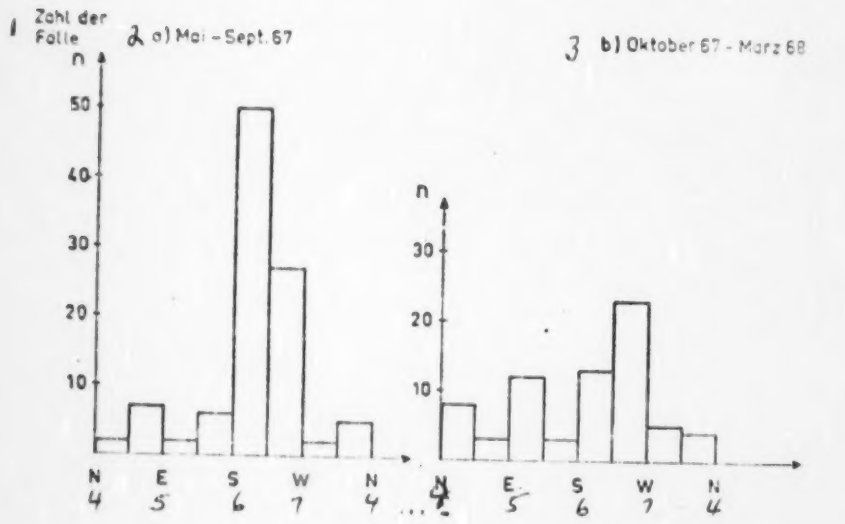


Illustration 7

Key:
1. Number of cases
2. May 1967 to September 1967
3. October 1967 to March 1968
4. North
5. East
6. South
7. West
8. Sampling measuring station Frankfurt/Main
9. Simultaneously occurring CO_2 and SO_2 concentration rises subject the wind direction

exhaust gas plume is subject to the wind velocity, that is, to the duration of the exhaust gases between the emission and immission in the atmosphere. With wind velocities of more than 2 m/sec., the values of the CO_2:SO_2 ratio lie within the range of approximately 200 - 700; with lesser wind velocities,

only values of more than 700 to a maximum of almost 3000 occur (Illustration 8). This relationship becomes clearer in Illustration 9. Here the individual cases with wind directions from the sector south to west have been subdivided once more. The simultaneous rises of the CO_2 and SO_2 concentrations in wind direction sector 180° to 225° were alloted to the power plant (with coal firing) at a distance of 2.5 km, and the rises in sector 225° to 270° to the power plant (mixed firing with coal and heating oil) at a distance of 10 km. The approximate duration of the exhaust gases can be calculated from the distance of the power plants and the wind velocity at the time of the concentration rise. On the ordinate in Illustration 9, the duration (or the transportation time) has been plotted and on the abscissa, the change of the CO_2:SO_2 ratio of the immission measurements relative to the value of this relation on the emission site.

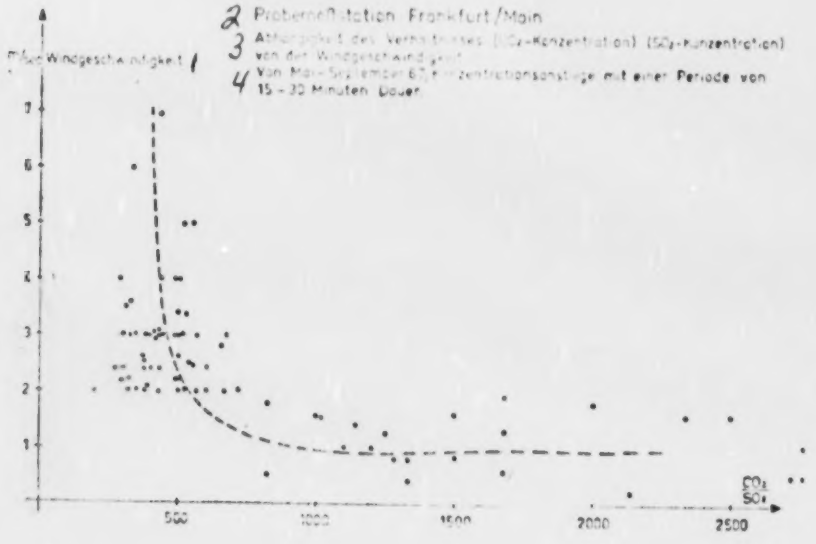


Illustration 8

Key: 1. Wind velocity
2. Sampling measuring station Frankfurt/Main
3. Ratio (CO_2 conentration):(SO_2 concentration) subject to wind velocity
4. Concentration rises from May to September 1967 with a period of 15 - 30 minutes duration

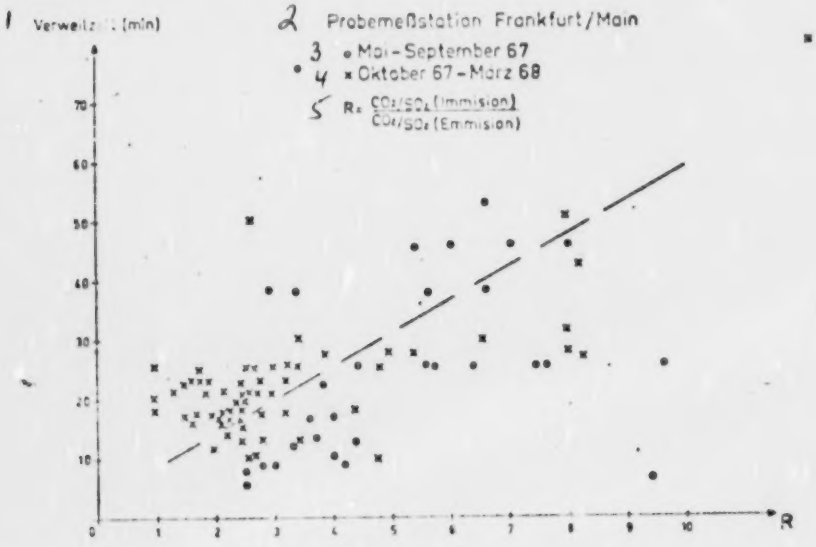


Illustration 9

Key: 1. Duration (min.)
2. Sampling measuring station Frankfurt/Main
3. May - September 1967
4. October 1967 - March 1968
5. $R = \frac{CO_2/SO_2 \text{ (Immission)}}{CO_2/SO_2 \text{ (Emission)}}$

The emission value of 200 was employed for the coal power plant, the value of 150 for the plant with the mixed firing [22]. It can be clearly recognized that the value of the ratio R (Illustration 9) rises with growing transportation duration: this, however, means an increasing SO_2 loss with ascending duration of the exhaust gases in the atmosphere. For evaluating the diffusion formulas, these tests can be of great significance. As a

rule, it is presupposed with the diffusion technique that the SO_2 concentration in a smoke plume does not alter through a chemical change to sulfates or through adsorption or absorption on the surfaces. Instead, it is assumed that a dilution of the exhaust gases occurs only through the comingling with atmospheric air. Should it be demonstrated that our results have general validity, then the computed ground concentrations under consideration of the additional SO_2 decrease should assume lower values.

During the winter months of October 1967 to March 1968, we examined the slow parallel changes of the CO_2 and SO_2 concentrations (Illustration 6). In these cases the influence of the low surface sources should be of special importance. Under the influence of decreasing horizontal wind velocity and increasing stabilization of the vertical interchange, the emissions from low chimneys of domestic fuel and motor vehicles can no longer be sufficiently removed [24]. The concentration of these emissions increases slowly in the whole agglomeration area, mostly over several hours. An influence from the large-scale emitters during such weather conditions is very unlikely since the emissions from high smoke stacks during a stable condition need a very long time in order to reach the ground again. The distance of the ground maximum is then often up to 50 km from the emitters. The slow increase of the concentration during the beginning stabilization of the air layer that is near the ground requires another type of evaluation of the recordings.

While during the short-period peaks of the immissions by the large-scale emitters (Illustration 5), we simply compared the level of the concentration increases with one another, here, at an interval of 15 minutes,

we ascertained the concentration increase of CO_2 and SO_2. It seemed reasonable to ascertain the average for each period out of all individual values and thus to calculate from this the characteristic CO_2:SO_2 ratio for the respective situation.

Again we represented these tests as frequency distribution:

1 Häufigkeitsverteilung des CO_2:SO_2-Verhältnisses
(Oktober 1967 bis März 1968)

2 Gruppe	3 Zentralstation	4 West- und Oststation
100—200	—	—
200—300	1,8	—
300—400	6,4	3,0
400—500	20,0	3,0
500—600	19,0	6,0
600—700	13,0	32,0
700—800	8,2	6,0
800—900	2,7	12,0
900—1000	2,7	15,0
1000—1100	—	6,0
1100—1200	0,9	6,0
1200—1300	—	3,0
5 Zahl der Fälle	(750)	(242)
6 Mittelwert	572	873

Table 10

Key: 1. Frequency distribution of the CO_2:SO_2 ratio, October 1967 - March 1968
2. Group
3. Central station
4. West and east station
5. Number of cases
6. Average

These values also are considerably higher than those indicated in Table 8 as characteristic for the emissions from small heating installations (coal, heating oil E). Also during this weather situation, evidently a considerable portion of sulfur dioxide is removed from the atmosphere. During

the winter with long-period rises, the averages only lie insignificantly higher than with the short-period peaks in the months outside of the heating period. According to Table 8, emissions from domestic fuel should show approximate CO_2:SO_2 values twice as high. The reason that the averages differentiate only very little is probably because of the shorter duration between the emission from the lower chimneys and the immission at the measuring site.

BREUER [25] found similar ratios with his experiments in Cologne and during a deep-lying inversion (December 1962) in Leverkusen. However, under the influence of a power plant, his values could not be verified here. The same thing is valid for a few measurements in Essen where BREUER had found values under the influence of domestic fuel which were in the range of 200 - 300. Strictly speaking, however, it is understandable that great differences must appear with just such tests since the duration of SO_2 in the atmosphere in the vicinity of emitters can be influenced through significant variable regional values.

The variability of the CO_2:SO_2 ratio and the missing consistency with the data of BREUER [25] leads to the realization that a direct conclusion cannot be drawn from the immission measurements of only such a test. However, we will continue the investigations in this direction since we are of the opinion that it will be possible to obtain data from the combined analysis of SO_2, CO_2, NO_2 and HC as well as from the meteorological parameter and the period of concentration changes, data that would show whether the immission concerns influences of large-scale emitters, room heater installations or other emitter groups.

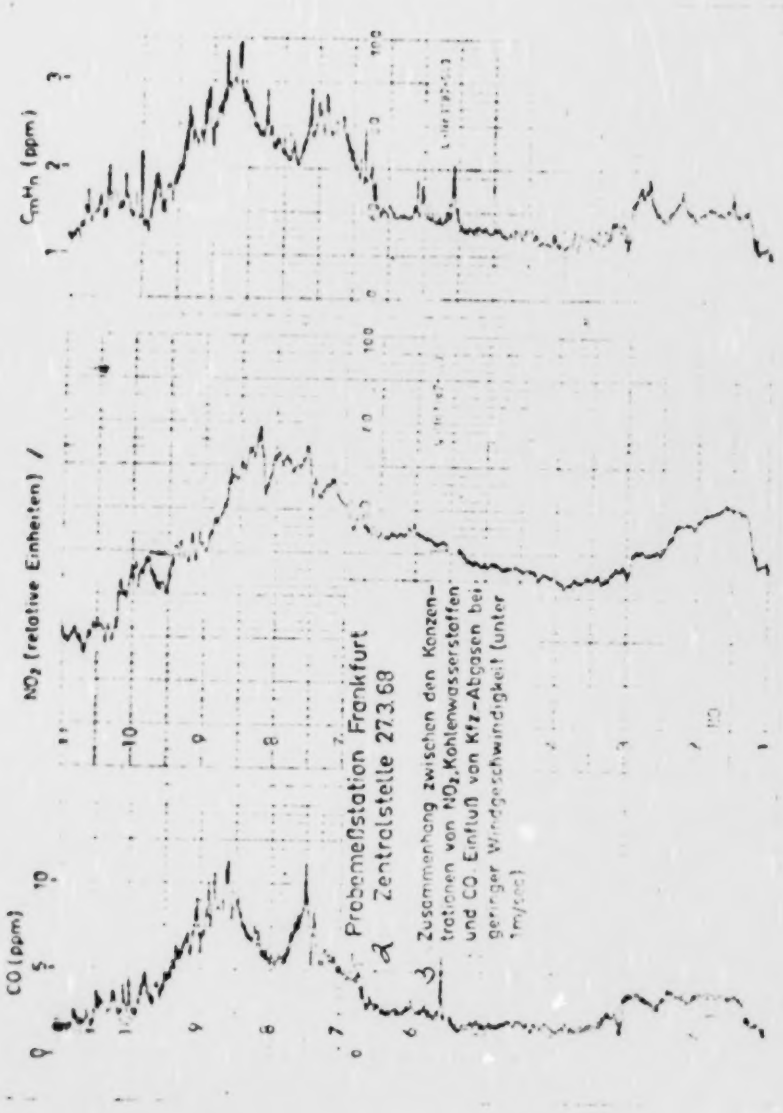

C_mH_n (ppm)
NO_2 (relative Einheiten)
CO (ppm)
Probemeßstation Frankfurt
Zentralstelle 27.3.68
3. Zusammenhang zwischen den Konzentrationen von NO_2, Kohlenwasserstoffen und CO. Einfluß von Kfz-Abgasen bei geringer Windgeschwindigkeit (unter 1 m/sec)

Illustration 10

Key: 1. Sampling measuring station Frankfurt
2. Central station 3-27-68
3. Interrelation between the concentrations of NO_2, hydrocarbons and CO. Influence of motor vehicle exhaust gases by slight wind velocities (under 1 m/sec.)

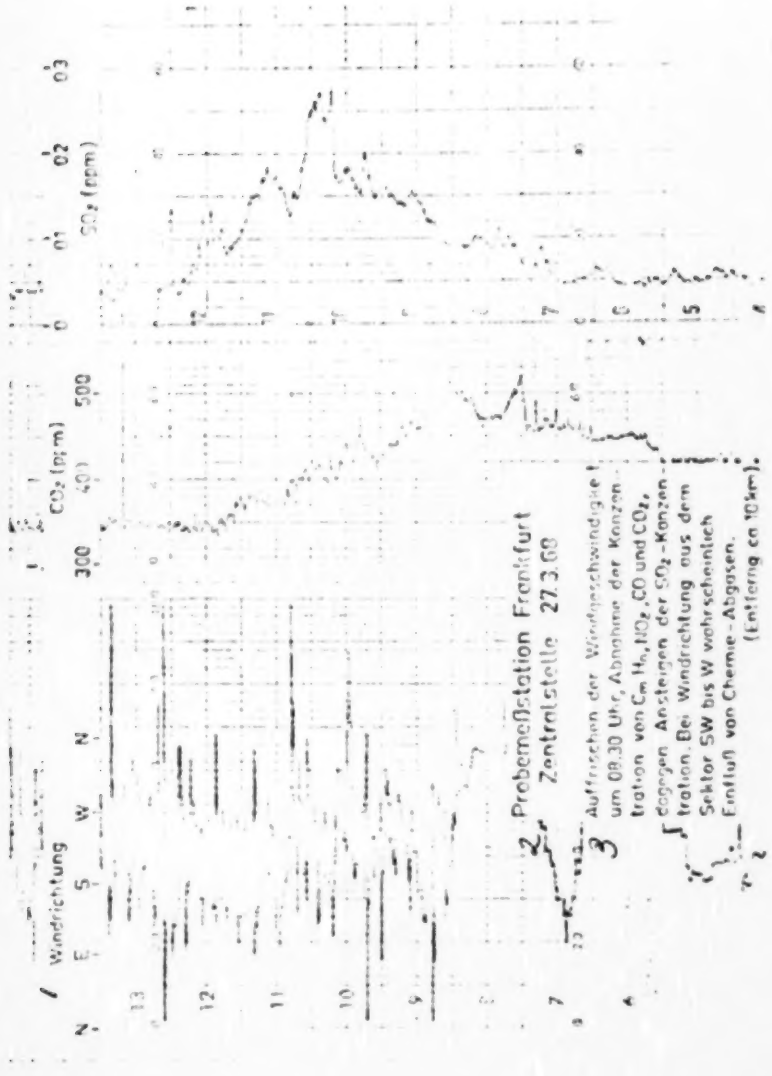

Windrichtung
N
W
S
E
N
SO₂ (ppm)
CO (mg/m)
2 Probemeßstation Frankfurt
Zentralstelle 27.3.68
3 Auffrischen der Windgeschwindigkeit um 08.30 Uhr, Abnahme der Konzentration von Cm Hn, NO₂, CO und CO₂, dagegen Ansteigen der SO₂-Konzentration. Bei Windrichtung aus dem Sektor SW bis W wahrscheinlich Einfluß von Chemie-Abgasen. (Entfernung ca 10 km).

Illustration 11

Key: 1. Wind direction
2. Sampling measuring station Frankfurt, central station 3-27-68
3. Regeneration of wind velocity around 8:30 a.m., decrease of C_m, H_n, NO_2, CO and CO_2, however, rise of the SO_2 concentration. With wind direction from the sector SW to W, probably influence of chemical exhaust gases (distance ca. 10 km)

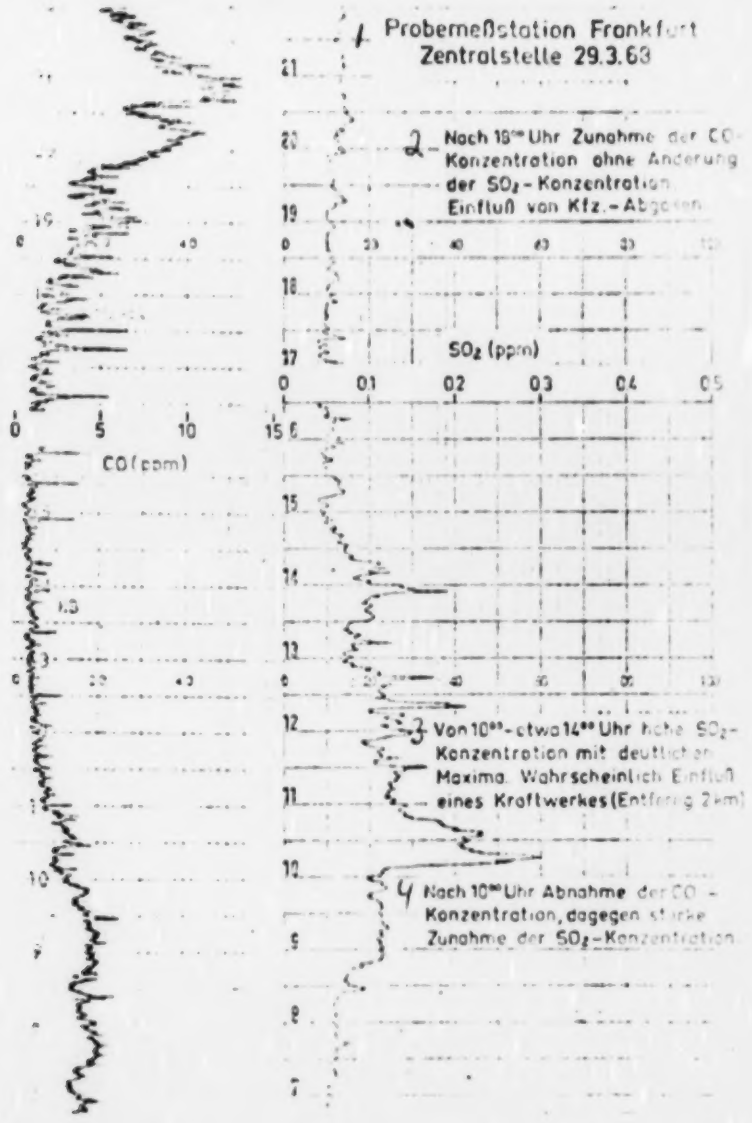

1 Probemeßstation Frankfurt
Zentralstelle 29.3.63
2 Nach 18^00 Uhr Zunahme der CO-Konzentration ohne Änderung der SO2-Konzentration. Einfluß von Kfz.-Abgasen
SO2 (ppm)
0 0,1 0,2 0,3 0,4 0,5
CO (ppm)
0 5 10
3 Von 10^00- etwa 14^00 Uhr hohe SO2-Konzentration mit deutlichen Maxima. Wahrscheinlich Einfluß eines Kraftwerkes (Entfernung 2 km)
4 Nach 10^00 Uhr Abnahme der CO-Konzentration, dagegen starke Zunahme der SO2-Konzentration

Illustration 12

Key: 1. Sampling measuring station Frankfurt, central station 3-29-68
2. After 6 p.m., increase of the CO concentration without change of the SO_2 concentration. Influence of motor vehicle exhaust gases
3. From 10:00 a.m. to approximately 2 p.m., high SO_2 concentration with distinct maxima. Probably influence of a power plant (distance 2 km)
4. After 10:00 a.m., decrease of the CO concentration, however, large increase of the SO_2 concentration

Besides characteristic components of the furnace exhaust gases (CO_2:SO_2), other emitter groups naturally can also be detected with the combination analysis. In Illustration 10, a case is demonstrated with the aid of the recording of the CO_2, NO_2 and HC concentration as it appears under the influence of motor vehicle exhaust gases and low wind velocity. Especially, the parallelism of the concentration course between the carbon monoxide and the total hydrocarbons (HC) is assured at almost every instant. Likewise, the course of the NO_2 concentration shows distinct parallels; however, the minimum of the CO and HC course is defined only very little there around 8 a.m.

An opposite motion of the CO_2 and SO_2 concentration can be recognized in Illustration 11. First, a concentration increase occurs until approximately 8:30 a.m.; then, under the influence of increasing wind velocity (here recognizable by the increasing gusts of the wind direction), the CO_2 concentration decreases almost linear until 12:00 p.m. However, the SO_2 concentration reaches its maximum around approximately 10:30 a.m.; only then does the decrease begin with a wind rotation from the west to west-southwest to southwest. Especially interesting in regard to this situation is the fact that the concentration change of SO_2 and CO_2 does not run parallel but at times in opposite direction. In this case, it does not deal with the influence of emissions from furnace installations. The maximum of the SO_2 concentration occurs with a wind direction from the sector around southwest. It is common knowledge that a sulfuric acid factory with a considerable SO_2 emission is located in that direction. It can be assumed with the aid of Illustration 11 that the exhaust gas plume of this smoke stack was observed at the measuring station.

Parallel diurnal variations of SO_2 and CO are represented in Illustration 12. A steep rise of the SO_2 concentration can be recognized in the morning after 10:00 a.m. and subsequently, additional maxima occur in irregular intervals until approximately 2 p.m. Through a comparison with the CO_2 concentration -- its representation has been dispensed with here -- the immissions could be assigned to the emissions of a power plant. During the evening hours, the CO concentration increases distinctly starting approximately at 6 p.m.; it reaches its maximum around 9 p.m. No change can be observed in the development of the SO_2 concentration. Therefore, it deals with the influence of motor vehicle exhaust gases during an interchange-poor weather condition. After 9 p.m., the CO concentration decreases again under the influence of the decreasing traffic density and reaches the normal concentration of approximately 1 ppm around 12:00 a.m., as it is usual for the traffic-poor night hours.

This representation is supposed to make especially clear that the development of the CO and SO_2 concentrations cannot exhibit any parallelism under certain conditions:

1. SO_2 immission under the influence of large-scale emitters
2. CO immission under the influence of motor vehicle exhaust gases during an interchange-poor weather condition and outside of the heating period.

During the heating period, case 2 will also always be associated with a rise in the SO_2 concentration. During 3-29-1968 (Illustration 12), the temperature around 6 p.m. amounted to 22° C; therefore, no emissions from house heating installations occurred. In comparison, case 1 will never be able to produce a rise in the CO concentration.

The following is an attempt to summarize the formulas of the combination analysis into individual points in one table:

1 Windgeschwindigkeit	2 Außentemperatur	3 Windrichtung	4 Auswirkung auf den Verlauf der Konzentration
5 niedrig	5 niedrig	–	8 Anstieg: SO_2, CO_2, CO, HC, NO_x
5 niedrig	6 hoch	–	8 Anstieg: CO, HC, NO_x
6 hoch	5 niedrig	7 vom Emittenten	8 Anstieg: SO_2, CO_2, (NO_x), (HC)
6 hoch	6 hoch	7 vom Emittenten	8 Anstieg: SO_2, CO_2, (NO_x), (HC)

Table 11

Key: 1. Wind velocity
2. Outside temperature
3. Wind direction
4. Effect on the course of the concentration
5. Low
6. High
7. From emitters
8. Rise

It can be recognized from this summarized grouping that, besides the analysis of the concentration rises, knowledge of meteorological diffusion conditions is necessary for the identification of the emitter. First of all, the decision has to be made whether the diffusion conditions will permit an enrichment from sources close to the ground or an influence of emissions from high-lying sources. The outside temperature gives additional information about the level of the emission from the room heater installations. Only after that should the measured gas-shaped components be assigned to an emitter group or, after consideration of the wind velocity and direction, to the individual emitter.

Unfortunately, with our present balance computer equipment, we cannot

undertake an evaluation on this type of EDV-equipment, which only permits the formation of half-hour averages. We are, therefore, limited to individual marked examples from the analogous recordings.

Summary

The first processing of measured values from a continuously recording station of this order, emperically is very time-consuming. Thus only after approximately five years after the start of the program did extensive reporting take place about the CAMP station [11]; likewise, the workings of the II. measuring program started in 1963 according to paragraph 7 of the Immission Protection Law of Nordheim-Westphalia [26] is now being published. We hope, however, that the first results of the sampling measuring station Frankfurt/Main, which are acquired on EDV-equipment, can be available approximately at the end of 1970. Later, the continuation of measuring and processing will be essentially more simple, since the preparation of the total data material, together with the already present programs for the computer equipment, can take place much faster.

The results, which were reported here, were essentially obtained without the use of an EDV. They show that the idea of such stations for monitoring the level of air pollution can be successful. The frequency distributions, for example, of sulfur dioxide, deviate only immaterially from the results of other measuring procedures (Table 3). Statements about the influence of different emitters can be made from the CO_2:SO_2 ratio, the wind velocity and direction. Simultaneously, the relative reduction of the SO_2 content during the transfer time through the atmosphere was determined. From combined analysis about the behavior of

other components, we could derive additional evidence about the influence of certain emitter groups on the composition of cutside air.

Especially, the results of the combination analysis represent an expansion of air-quality research. It is possible through this to determine the degree and duration of the influence on the quality of the air in an area around the measuring station by certain emitters or emitter groups. Simultaneously, we can estimate which improvements can be expected through the reduction of the ejection of certain emitters or emitter groups. Thus it can be avoided that measures for the emission reduction are applied at the wrong place and thus without remarkable effect. Through the evaluation of future developments, supplementary information is gained and taken into account. These considerations are especially suitable for the use of planning and sanitation measures within the scope of the national planning law. Especially here it is important to achieve the best results with the least means. Measurements of the sampling measuring station or similar systems give data about the following questions:

1. which air pollution is already in existence and during which weather conditions do increased concentrations occur,

2. which emitters participate,

3. how are these emissions developing in the course of the next years and what effect does this have on the level of the immission?

It still seems questionable whether direct measures can be initiated against a certain emitter on ground of the measurements at the sampling measuring station.

This will only be possible when the emitter is known, if the

immissions can be assigned unobjectionally and if exceeding the permissable concentration was measured. In addition, it must be taken into consideration that the measuring station must not necessarily be located at the site of the highest ground concentration. For reasons of statistical analysis about meteorological conditions (for example, diffusion classes and wind direction), it would be conceivable that improvements could be carried out with the aid of theoretical considerations without exceeding the threshold at the measuring station.

It would be desirable for the future to still expand the sampling measuring station through a station for detecting the car exhaust gases at a place of traffic density and a station located outside of the city in a residential area. With this, the recording and processing of the measured values should be changed simultaneously. At this time, only averages are produced; these are printed and are only later evaluated in an EDV-installation. It should, however, be our goal to have fast, usable results available for air quality questions. For many purposes it would be quite uninteresting, perhaps to be only able to say after one or two weeks why, for example, with an inversion situation certain diseases or annoyances or even increased death rates occurred.

In order to have very quick results available, computations should take place in a so-called "On-Line Real-Time System." Such a data processing system is already in use in Chicago [27]. The measured values are fed by punched cards or such into the computer unit without interpolation; besides storing the measured values, the system carries out computation operations and merely prints out the computed results. All measured values and intermediate results are stored on magnetic tapes. If such a

small computer installation is programmed especially for air quality purposes, then all desired data can be withdrawn at any time, for example, the dose of a substance during the last 48 hours, or the present horizontal distribution of the concentration in the total monitoring area.

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